



APPEAL BRIEF

Before

Board of Patent Appeals and Interferences

in the

United States Patent and Trademark Office

#18
8/16/00

Ex Parte

U.S. Serial No: 08/986,696

Inventor: Moses O. Jejelowo et al.

Title: Polymerization Catalyst Systems and their Use

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I. REAL PARTY IN INTEREST

Univation Technologies



II. RELATED APPEALS AND INTERFERENCES

No pending appeals or interferences are known to Appellants

III. STATUS OF CLAIMS

In the Final Office Action mailed March 20, 2000 pending claims were 10-14, 16-23, 25-32 and 51-57. Claims 10-14, 16-23, 25-32 and 51-57 were rejected.

The Final Rejection of claims 10-14, 16-23, 25-32 and 51-57 is appealed.

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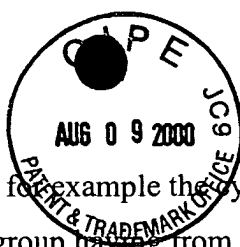
IV. STATUS OF AMENDMENTS

In the Advisory action mailed June 13, 200 in response to Appellants' Amendment under 37 CFR 1.116 filed May 26, 2000 the Examiner indicated that the amendment would be entered upon filing a Notice of Appeal and an Appeal Brief. In the amendment, claims 10, 12, 13, 20, 21, 28, 52, 53 and 54 were amended. The Examiner has indicated that upon entry of the amendment, the rejections under 35 U.S.C. § 112 would be withdrawn.

V. SUMMARY OF THE INVENTION

A problem which existed in the prior art was the absence of a polymerization process employing a hafnocene based catalyst system that would produce a high molecular weight polyolefin, and do so with high activity. The prior art hafnocene based catalysts did produce high molecular weight polyolefins but did so with poor polymerization activity (page 1, line 34 carried over to page 2, line 6).

The claimed inventions are directed to a process of polymerizing olefins in the presence of a catalyst system comprising a hafnium metallocene and an activator wherein



at least one ligand, for example the cyclopentadienyl ring, is substituted with at least one linear or iso alkyl group having from 3 to 10 carbon atoms (page 2, lines 20 – 24; page 3, lines 13 – 16; page 7, line 29; page 8, lines 3 and 4). The activators are defined at page 9, line 10 carried over to page 10, line 3. Supports are defined at page 11, lines 4 – 26. Independent claim 21 is directed toward a continuous gas phase process in a fluidized bed gas phase reactor (Page 15, lines 7 – 35). Independent claim 28 is directed toward a continuous slurry phase process (page 16, lines 7 – 31)

VI. ISSUES

The issues before the Honorable Board of Patent Appeals and Interferences are:

Whether the process claimed in claims 10 to 12, 14, 16, 19, 28 to 30, 51, 52, 55 and 56 is obvious under 35 U.S.C. § 103 (a) over U.S. Patent No. 5,621,054 of Harrington.

Whether the process claimed in claims 10 to 14, 16, 17, 20, 27 to 32, 51 to 54, 56 and 57 is obvious under 35 U.S.C. § 103 (a) over U.S. Patent No. 5,387,660 of Doyle et al. (Doyle).

Whether the process claimed in claims 10 to 14, 16 to 23, 25, 26, 28 to 31 and 51 to 57 is obvious under 35 U.S.C. § 103 (a) over U.S. Patent No. 5,281,679 of Jejelowo et al. (Jejelowo).

Whether the process claimed in claims 18, 19, 21 to 23, 25 and 26 is obvious under 35 U.S.C. § 103 (a) over Doyle optionally in view of U.S Patent No. 5,714,426 of Tsutsui et al. (Tsutsui).

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VII. GROUPING OF CLAIMS



The claims before the Honorable Board of Appeals and Interferences stand together.

VIII. ARGUMENTS

As discussed in Applicants' response mailed January 11, 2000, it is well known in the art that polymerization processes employing hafnocene based catalyst systems obtain high molecular weight polymer product, but manifest relatively poor activity as compared with the titanocene and zirconocene catalyst systems. There has been a long felt need for a polymerization process employing a hafnocene catalyst system that would not only obtain the much desired high molecular weight polymer product, particularly polyolefins, but would do so in accordance with a process that manifest high activity. In accordance with Applicants' invention it has been discovered that hafnocene based catalyst systems containing the claimed alkyl substituents on the cyclopentadienyl ring manifest highly improved activity. A comparison of Applicants' claimed process with polymerization processes comprising hafnocenes catalysts absent the alkyl groups which are recited in the instant claims evidences unexpected results. It is respectfully believed there is nothing in the art of record that would lead one of ordinary skill to predict that by placing the claimed alkyl groups on the ligands of hafnocene catalyst that the catalytic activity would greatly increase. Table 1 found on page 24 of the application shows polymerization processes comprising the catalysts in accordance with the claimed invention obtain activities that are about 50 times greater and yields that are about 10 to 20 times greater as compared with hafnocenes based catalysts absent the alkyl substituents which are recited in the claims before the Honorable Board. The unexpectedness and unobviousness is all the clearer when one notes that the improved activity and yields were obtained without any loss in the high molecular weight range.

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THE REJECTION OVER U.S. PATENT NO. 5,621,054

Claims 10 to 12, 14, 16, 19, 28 to 30, 51, 52, 55 and 56 have been finally rejected as being obvious under 35 U.S.C. § 103 (a) over U.S. Patent No. 5,621,054 of Harrington.

As discussed above, it is submitted that the unobviousness with respect to the invention is the fact that a hafnocene catalyst system has been discovered which manifests high activity. It is submitted that the examples in Harrington support the unobviousness of the claimed invention. When one examines the working examples of Harrington one observes that the activity, as determined by the yield and reaction time, for the zirconocenes and titanocenes is much greater than the activity, for the hafnocenes (Table 1, examples 4 to 8 vs. 2, 3, 9 and 10). The hafnocene activity in each of examples 2, 3, 9 and 10 is very low. In example 1, the yield was high however: the reaction time was higher than in examples 2, 3, 9 and 10; the amount of norbornene employed was higher than in the remaining examples; and the amount of catalyst employed was greater than the examples employing the zirconocene and titanocene or the remaining hafnocene catalysts. From Harrington alone when combining Table 1 and 2 one of ordinary skill in the art learns that hafnocenes with respect to both activity and MW would not be a catalyst of choice.

The Examples in the instant application unexpectedly demonstrate a very high catalytic activity. From the teachings and disclosure of Harrington, one of ordinary skill in the art could not predict the results as unequivocally demonstrated in the instant application.

The Examiner urges that Harrington by exemplifying t-butyl has particularly pointed to alkyl substituents of the size corresponding to those instantly claimed and that common alkyl groups are satisfactory for use. Contrary to the Examiner's suggestion, no where within the four corners of Harrington is there any statement wherein the "patentee clearly states that common alkyl groups are satisfactory for use at the required substitution site." Harrington does state that "any C₁ or higher carbon number or alkyl

substituted silyl substituent having at least one carbon atom can be used” The patentee then teaches that disubstituted substitution patterns can be employed as well as alkenyl, aryl, alkylaryl, or aryalkyl radicals. Many more substitutions are described in Col. 4 lines 16 – 32 cited by the Examiner. In any event, Harrington is describing substituents for zirconocenes, titanocenes as well as hafnocenes without any distinction. Furthermore upon combining Harrington’s examples together with the Examiner’s reasoning one can only conclude that alkyl substituents close in size to t-butyl (substituent employed in Patentee’s examples) do not obtain high activity for hafnocenes. The Examiner’s own reasoning when examining the evidence in the Harrington would teach away from a polymerization process employing the hafnocenes recited in the claims before the Honorable Board.

It is respectfully submitted that the Examiner has arrived at a conclusion without supportive evidence. Harrington does not single out the t-butyl substituent but in fact states that the tert-butyl substituted hafnocene ... “exemplifies the class of compounds suitable in this invention” (Col. 3, lines 66 and 67) and then goes on to list all the substituents available in the Col. and lines cited by the Examiner. The conclusion that, “One of ordinary skill in the art would be motivated to use simple linear or branched alkyl substituents of common $C_3 - C_{10}$ linear or iso alkyl group, because Harrington has stated that such use results in effective polymerization” has not been supported by any evidence excepting the Examiner’s own position. The erroneous conclusion is easily recognized when one reads all the possible substituents found in Col. 4, lines 16 - 32. Why one of ordinary skill in the art would be particularly led to applicant’s claimed substituents in view of the multitude of substituents disclosed by the examiner has not been explained. It has been respectfully requested that the Examiner present evidence outside of his own opinion that would support the specific selection from the multitude of substituents presented in the Col. and line cited by the Examiner. In addition to not listing a single metallocene having a linear or iso alkyl substituent on the Cp ring, Harrington does not address activity. It is respectfully submitted that there is nothing in Harrington that would motivate the ordinary practitioner in the art to substitute Appellants’ recited substituents for any of the specific substituents with any expectation

of success in obtaining a high activity hafnocene catalyst system. For that matter there is no motivation provided in Harrington to research for a high activity polymerization process employing any hafnocene catalyst. There is, it is respectfully submitted, not even an inference from which one skilled in the art would be led to Appellants' process and predict or expect such a process would manifest high activity.

In view of the above, it is respectfully submitted that the Examiner has not established a prima facie case of obviousness. It is further submitted that the Examples in the instant case do demonstrate unexpected results. Appellants have unequivocally demonstrated that the class of substituents recited in the instant claims obtains activities that are about 40 – 50 times greater than the hafnocene catalysts comprising substituents outside the claimed substituents.

In view of the above it is respectfully asked that the Examiner's Final rejection over Harrington be reversed.

THE REJECTION OVER U.S. PATENT NO. 5,387,660

Claims 10 to 14, 16, 17, 20, 27 to 32, 51 to 54, 56 and 57 have been finally rejected under 35 U.S.C. § 103(a) as being unpatentable over Doyle et al. ("Doyle"). This rejection is respectfully traversed. As in Harrington discussed above, Examiner arrives at a very specific conclusion from a very broad disclosure. The Examiner should not only recite Col. 3, lines 51-53, but should include line 51-60. By reading the complete Doyle disclosure one notes that the only group specially recited in the specification other than in the working examples is "methyl". The patentee at Col. 3, lines 58 specifically refers to "pentamethylcyclopentadienyl groups as giving very good results but only in respect to Patentee's process which is directed to the addition of more catalyst component when the process activity decreases. The Examiner errs in stating that "the working examples use t-butyl substituent." Only examples 6 and 7 do. Examples 1 to 5 employ methyl substituents on the cyclopentadienyl ring and Example 5 is drawn to a hafnocene.

It is respectfully submitted that the evidence Examiner relies upon is absent a reliance on logic and sound scientific principles. Merely because there are "15" possible structural isomers corresponding to C1-C5 alkyl groups some of which would include iso and linear group is not evidence. Based on the total disclosure by Doyle it is urged that that the number of possible isomers would no more suggest Appellants' claimed substituents than it would suggest substituents outside of that recited in the instant claims. Based on the disclosure at Col. 3, line 58 one would at most be lead to the use of methyl groups rather than larger alkyl group. Certainly there is no motivation in Doyle to employ a process comprising the catalysts recited in the claims before the Honorable Board with the expectation of obtaining a highly active polymerization process. The attempt at providing motivation is at most weak. The Examiner again reaches a conclusion from that which does not appear in Doyle. The patent does not recite iso or linear and does not exemplify or list iso or linear groups. The Examiner errs in stating that Doyle "has set forth a group of substituents which substantially overlaps with the group specified in the instant claims." Appellants have on the other hand provided data which distinguishes the claims containing the iso and linear limitations over other radicals.

Again, the Honorable Board is referred to the Examples in Appellants' application. The Examples that cover catalysts within the scope of the claims demonstrate superior activity over the examples, which recite catalysts outside the scope of the claims. Please note that the comparison examples in this case employ a methyl group, the only named group in Col. 3, lines 50 to 60 of Doyle. In view of the above, it is respectfully submitted that the Examiner's final rejection over Doyle should be reversed.

THE REJECTION OVER U.S. PATENT NO. 5,281,679

Claims 10 to 14, 16 to 23, 25, 26, 28 to 31 and 51 to 57 have been rejected under 35 U.S.C. §103 (a) as being unpatentable over Jejelowo et al. (Jejelowo)

It is respectfully submitted that one of ordinary skill in the art would take Official Notice (it is very well known in the art) that the hafnocene catalysts (prior to Applicants' invention) produced high molecular weight polymer product but did so at a very slow catalytic activity. The Examples in Appellants' patent application are contrary to that which would be understood by the ordinary practitioner in the art. It is respectfully submitted that the ordinary practitioner would neither predict nor expect the substantial increase in catalytic activity obtained in accordance with Appellants' claimed process and demonstrated in the Example over the catalysts not within the scope of the claimed invention. Appellants have demonstrated criticality with respect to the recited substituents for hafnocenes.

The Examiner cites MPEP 2144 and states "that a reference must suggest desirability of a substitution for the same reason as applicants' purpose has no legal basis." It is respectfully submitted that the MPEP also states that if the applicants have demonstrated criticality of a specific limitation, it would not be appropriate to rely solely on case law as the rationale to support an obviousness rejection. Appellants have shown criticality for the claimed process employing the recited hafnocenes.

The numerous cases the Examiner alludes to as discussed in MPEP 2144 do not hold that a broad, indefinite disclosure makes obvious a specific, definite recitation of a claimed invention. Jejelowo provides a broad disclosure for Patentee's purposes, i.e. to broaden the molecular weight distribution and enhance tensile impact strength of polymer product. The skilled practitioner in the art if researching for a process of improving the hafnocene catalytic activity would not find motivation in Jejelowo to employ a polymerization process using the hafnocenes as recited in the claims before the Honorable Board. Jejelowo furthermore supports the unobviousness of Appellants' invention. The Examiner has pointed to the Examples at Col. 17 and 18. Please note that the examples having the substituents that fall within the definition of Appellants' substituents (but are not hafnocenes) evidence lower activity than the remaining examples. For example, a compound using tBu obtains a catalyst rate of 0.671, 1.001 and 0.921 whereas the compounds using iPr obtains catalyst rates of 0.158, 0.225, 0.233. A

skilled practitioner in the art reading such a disclosure would naturally expect to obtain low activities and not be motivated to Appellants' substituents. In view of the above, it is respectfully submitted that the Examiner's final rejection over Jejelowo should be reversed.

THE REJECTION OVER U.S. PATENT NOS. 5,387,660 and 5,714,426

Claims 18, 19, 21-23, 25 and 26 have been rejected as unpatentable over 103 & S. Patent 5,387,660 to Doyle et al., optionally in view of U.S. Patent 5,714,426 to Tsutsui et al. 35 U.S.C. § 103(a).

The Examiner states that Doyle et al. discuss all aspects of the claimed invention except for the use of a supported catalyst and the use of gas-phase conditions.

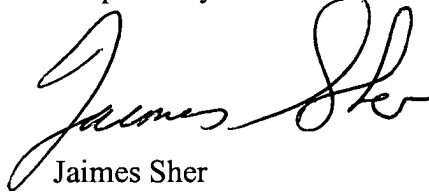
For reasons discussed above it is submitted that, contrary to the Examiner's statement, no where within the four corners of Doyle is there any discussion discussing all aspects of the Appellants' claimed invention. Doyle does not discuss nor make obvious in the sense of 35 U.S.C. 103 increasing the catalytic activity of hafnocenes. Doyle does not suggest that by use of substituents recited in the instant claims that one can improve the catalytic activity of a polymerization process. Doyle does not mention a single substituent in the general disclosure except for methyl. In the examples of Doyle the only substituents employed are methyl and t-butyl, both being outside the scope of Appellants' claimed invention. Tsutsui does not solve the deficiencies in Doyle. At most, one would employ the process conditions of Tsutsui with the catalyst systems of Doyle. In so doing it is respectfully submitted that there would be no motivation for a practitioner in the art to employ Appellants' claimed process. It is respectfully submitted that a skilled practitioner would find Appellants' results as illustrated in the application examples unexpected and unobvious. In view of the above, it is respectfully submitted that the Examiner's final rejection over Doyle, optionally in view of Tsutsui should be reversed.

SUMMARY

In summary, there is no teaching or suggestion in the references cited by the Examiner either alone or combined which make obvious Appellants' claimed process. Particularly, there is no teaching or suggestion in the references cited by the Examiner either alone or combined which makes obvious Appellants' claimed process in order to provide a high activity hafnocene catalytic polymerization process obtaining a high molecular weight polymer product.

In view of the above it is respectfully submitted that the all the rejections in this appeal should be reversed.

Respectfully submitted



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IX. APPENDIX

Claims 10-14, 16-23, 25-32 and 51 -57 involved in the appeal are listed below.

10. A process for polymerizing olefin(s) in the presence of a catalyst system comprising a hafnium metallocene catalyst compound having at least one ligand substituted with at least one linear or iso alkyl group having from 3 to 10 carbon atoms, and an activator.

11. Th process in accordance with claim 10 where the linear or iso alkyl group has 3 to 5 carbon atoms.

12. The process in accordance with claim 10 wherein the ligand is one or more cyclopentadienyl ligands, wherein one of the cyclopentadienyl ligands is substituted with at least one linear or iso alkyl having 3 or 4 carbon atoms.

13. The process in accordance with claim 10 wherein the ligands are two cyclopentadienyl rings each substituted with at least one linear or iso alkyl group having 3 – 10 carbon atoms.

14. The process in accordance with claim 11 wherein the linear or iso alkyl group is selected from one or more of the group consisting of n-propyl, isopropyl, n-butyl, isobutyl and n-pentyl.

16. The process in accordance with claim 10 wherein the olefin(s) are alpha-olefins having from 2 to 12 carbon atoms.

17. The process in accordance with claim 10 wherein the olefin(s) are ethylene in combination with one or more other alpha-olefins having from 3 to 10 carbon atoms.

18. The process in accordance with claim 10 wherein the process is a gas phase process.

19. The process in accordance with claim 10 wherein the catalyst system further comprises a support.

20. The process in accordance claim 10 wherein the hafnium metallocene catalyst compound is bis(n-propyl-cyclopentadienyl) hafnium dichloride.

21. A continuous gas phase process for polymerizing olefin(s) in a fluidized bed gas phase reactor in the presence of a catalyst system to produce a polymer product, the catalyst system comprising a ligand hafnium metallocene catalyst compound having at least one ligand substituted with at least one linear or iso alkyl group having from 3 to 10 carbon atoms, and the polymer product comprising less than 2 ppm hafnium.

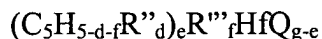
22. The process of claim 21 wherein the polymer product comprises less than 1 ppm hafnium.

23. The process in accordance with claim 21 wherein the catalyst system is supported.

25. The process of claim 21 wherein the polymer product wherein the polymer product has a density greater than 0.915 g/cc.

26. The process in accordance with claim 21 wherein the olefin(s) are ethylene and at lest one alpha-olefin having 3 to 8 carbon atoms.

27. The process in accordance with claim 21 wherein the catalyst system is represented by the formula:



wherein $(C_5H_{5-6-f}R''_d)$ is an unsubstituted or substituted cyclopentadienyl ligand bonded to Hf, wherein at least one $(C_5H_{5-6-f}R''_d)$ is substituted with at least one R'' which is an alkyl group selected from the group consisting of n-propyl, isopropyl, isobutyl and n-pentyl, each additional R'' , which can be the same or different is hydrogen or a substituted or unsubstituted hydrocarbyl having from 1 to 30 carbon atoms or combinations thereof or two or more carbon atoms are joined together to form a part of a substituted or unsubstituted ring or ring system having 4 to 30 carbon atoms, R''' is one or more or a combination of the group consisting of carbon, germanium, silicon, phosphorous and nitrogen atoms containing radical bridging two $(C_5H_{5-6-f}R''_d)$ rings, or bridging one $(C_5H_{5-6-f}R''_d)$ ring to Hf; each Q which can be the same or different is selected from the group consisting of hydride, substituted and unsubstituted hydrocarbyl having from 1 to 30 carbon atoms, halogen, alkoxides, aryloxides, amides, phosphides and combination thereof; two Q's together form an alkylidene ligand or cyclometallated hydrocarbyl ligand or other divalent anionic chelating ligand; where g is an integer corresponding to the formal oxidation state of Hf, d is 0, 1, 2, 3, 4, or 5, f is 0 or 1 and e is 1, 2, or 3, and the polymer product has a melt index less than 0.1 dg/min (ASTM D-1238-F or ASTM D-1238-E) without the addition of hydrogen to the process.

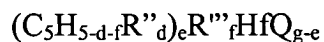
28. A continuous slurry phase process for polymerizing olefin(s) in the presence of a catalyst system to produce a polymer product in a liquid polymerization medium, the catalyst system comprising a hafnium metallocene catalyst compound having at least one substituted with at least one linear or iso alkyl group having from 3 to 10 carbon atoms, and the polymer product comprising less than 2 ppm hafnium.

29. The process in accordance with claim 28 wherein the polymer product comprises less than 1 ppm hafnium.

30. The process in accordance with claim 28 wherein the density is greater than 0.900 g/cc.

31. The process in accordance with claim 28 wherein the olefins are ethylene and at least one alpha-olefin having 3 to 8 carbon atoms.

32. The process in accordance with claim 28 wherein the catalyst system is represented by the formula:



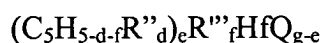
wherein $(C_5H_{5-d-f}R''_d)$ is an unsubstituted or substituted cyclopentadienyl ligand bonded to Hf, wherein at least one $(C_5H_{5-d-f}R''_d)$ is substituted with at least one R'' which is an alkyl substituent having 3 or more carbon atoms, each additional R'' , which can be the same or different is hydrogen or a substituted or unsubstituted hydrocarbyl having from 1 to 30 carbon atoms or combinations thereof or two or more carbon atoms are joined together to form a part of a substituted or unsubstituted ring or ring system having 4 to 30 carbon atoms, R''' is one or more or a combination of the group consisting of carbon, germanium, silicon, phosphorous and nitrogen atoms containing radical bridging two $(C_5H_{5-d-f}R''_d)$ rings, or bridging one $(C_5H_{5-d-f}R''_d)$ ring to Hf; each Q which can be the same or different is selected from the group consisting of hydride, substituted and unsubstituted hydrocarbyl having from 1 to 30 carbon atoms, halogen, alkoxides, aryloxides, amides, phosphides and combination thereof; two Q's together form an alkylidene ligand or cyclometallated hydrocarbyl ligand or other divalent anionic chelating ligand; where g is an integer corresponding to the formal oxidation state of Hf, d is 0, 1, 2, 3, 4, or 5, f is 0 or 1 and e is 1, 2, or 3, and the polymer product has a melt index less than 0.1 dg/min (ASTM D-1238-F or ASTM D-1238-E) without the addition of hydrogen to the process.

51. The process in accordance with claim 12 wherein the linear or iso alkyl group has 3 carbon atoms.

52. A process for polymerizing olefin(s) in the presence of a catalyst system comprising a hafnium transition metal metallocene catalyst having at least one

cyclopentadienyl ring substituted with at least one alkyl group selected from group consisting of n-propyl, isopropyl, isobutyl and n-pentyl, and an activator.

53. A continuous gas phase process for polymerizing olefin(s) in a fluidized bed gas phase reactor in the presence of a catalyst system to produce a polymer product, the catalyst system comprising a bulky ligand hafnium transition metal metallocene catalyst represented by the formula:



wherein $(C_5H_{5-d-f}R''_d)$ is an unsubstituted or substituted cyclopentadienyl ligand bonded to Hf, wherein at least one $(C_5H_{5-d-f}R''_d)$ is substituted with at least one R'' which is an alkyl group selected from the group consisting of n-propyl, isopropyl, isobutyl and n-pentyl, each additional R'' , which can be the same or different is hydrogen or a substituted or unsubstituted hydrocarbyl having from 1 to 30 carbon atoms or combinations thereof or two or more carbon atoms are joined together to form a part of a substituted or unsubstituted ring or ring system having 4 to 30 carbon atoms, R''' is one or more or a combination of the group consisting of carbon, germanium, silicon, phosphorous and nitrogen atoms containing radical bridging two $(C_5H_{5-d-f}R''_d)$ rings, or bridging one $(C_5H_{5-d-f}R''_d)$ ring to Hf; each Q which can be the same or different is selected from the group consisting of hydride, substituted and unsubstituted hydrocarbyl having from 1 to 30 carbon atoms, halogen, alkoxides, aryloxides, amides, phosphides and combination thereof; two Q's together form an alkylidene ligand or cyclometallated hydrocarbyl ligand or other divalent anionic chelating ligand; where g is an integer corresponding to the formal oxidation state of Hf, d is 0, 1, 2, 3, 4, or 5, f is 0 or 1 and e is 1, 2, or 3, and the polymer product has a melt index less than 10 dg/min (ASTM D-1238-F or ASTM D-1238-E) without the addition of hydrogen to the process.

54. The process in accordance with claim 10 wherein the hafnium metallocene catalyst compound is a bis-cyclopentadienyl hafnium metallocene compound.

55. The process in accordance with claim 10 wherein the activator is alumoxane.

56. The process in accordance with claim 13 where in the linear or iso alkyl group has from 3 to 5 carbon atoms.

57. The process in accordance with claim 10 wherein the linear or iso alkyl group has 3 carbon atoms.